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DOSSIER

DU COMBUSTIBLE NUCLÉAIRE AUX DÉCHETS : RECHERCHES ACTUELLES

FROM NUCLEAR FUELS TO WASTE: CURRENT RESEARCH

Plutonium and minor actinides management in the nuclear fuel cycle: assessing and controlling the inventory

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Abstract The mastering of the plutonium and minor actinides inventory in the French Nuclear Cycle is based on a progressive approach from the present status, dealing with the partial reprocessing of spent fuels and the recycling of Pu in the MOX assemblies loaded in the 20 licensed PWRs. This strategy keeps the door open long-term, for example, for the eventual multirecycling of excess Pu in dedicated new assemblies, such as APA or CORAIL in order to stabilise the Pu inventory in the fuel cycle or allow its utilization in new types of fast reactors. Presently, in the framework of 1991 law, scenario studies relying on present and/or innovative technologies are carried out in order to transmute both Pu and minor actinides, thus minimising the quantities to be for disposal. ***To cite this article: H. Mouney, C. R. Physique 3 (2002) 773–782.***

back-end of fuel cycle / plutonium / minor actinides / transmutation

La problématique de la maîtrise de l'inventaire plutonium et actinides mineurs dans le cycle du combustible nucléaire

Résumé La maîtrise de l'inventaire en plutonium et actinides mineurs dans le cycle du combustible nucléaire du parc de réacteurs français s'appuie sur une démarche progressive à partir de la politique actuelle de retraitement partiel du combustible usé et du recyclage du plutonium séparé, dans les combustibles MOX, chargés dans les 20 réacteurs REP qui en ont reçu l'autorisation. Cette stratégie laisse la porte ouverte pour le long terme, par exemple pour le multirecyclage éventuel du plutonium excédentaire dans de nouveaux concepts d'assemblages, tels que APA ou CORAIL, afin d'en stabiliser l'inventaire dans le cycle ou pour son utilisation dans de nouvelles générations de réacteurs rapides. Enfin, dans le cadre de la loi de 1991, des scénarios mettant en œuvre des réacteurs conventionnels et/ou innovants sont étudiés afin de transmuter les actinides mineurs, pour en réduire les quantités résiduelles dans les déchets ultimes destinés au stockage. ***Pour citer cet article : H. Mouney, C. R. Physique 3 (2002) 773–782.***

aval du cycle / plutonium / actinides mineurs / transmutation

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1. Introduction: the current management of EDF's nuclear power facilities

The current management of the fuel cycle is to reprocess about 850 metric tons per year of spent UOX fuel, out of the 1 150 metric tons (UOX + MOX) unloaded per year from the PWR reactors of EDF's nuclear power facilities. UOX fuel is made of UO_2 ceramic pellets, U being enriched in U235, MOX fuel is made of mixed UO_2–PuO_2.

This reprocessing allows EDF to recover yearly approximately 8.5 metric tons of plutonium which are used for the production of 100 metric tons of MOX fuel loaded into the 20 PWR units ('REP 900') authorised to recycle this fuel in their core up to a rate of 30% of the overall fuel.

From an initial average burn-up fraction of 33 $GWd{\cdot}t^{-1}$, the fuel used in EDF reactors have reached, over the last years, 44 $GWd{\cdot}t^{-1}$ for UOX and about 37 $GWd{\cdot}t^{-1}$ for MOX fuel, the use of which is more recent.

In the coming years, a significant increase in the burn-up fraction of UOX fuel is scheduled, which will improve the energy performance of the fuel. Considered from the back-end of the fuel cycle, this will result in a reduction of the number of unloaded spent fuel assemblies [1].

The coming implementation of the MOX Parity management will serve to balance the reprocessing and recycling fluxes and to limit the stock of separated plutonium to 3 years of MOX fuel production.

All of the spent fuel unloaded from the NPPs is sent to the La Hague plant for reprocessing. The capability of MOX to be reprocessed has been proven, even though its processing is not yet scheduled in the short term by EDF.

The storage pool capacity for spent fuel providing the required safety level is sufficient in the coming 20–30 years and especially according to the forecasts of an increase in irradiation of the UOX assemblies. Assuming a constant reprocessing rate (850 $t{\cdot}year^{-1}$), this will lead to a decrease in the UOX quantities stored in view of their reprocessing as of 2015 (12 000 t by that time). The actual storage time of the UOX and MOX fuel before reprocessing may vary and allows one to postpone the decisions to be taken later. In this respect, the development or not of reactors capable of burning minor actinides will be decisive in stating precisely the storage time of these fuel types, their elimination and final destination.

2. Increased MOX performance

On unloading, the currently spent UOX fuel reach, after 4 cycles of irradiation, a discharge burn-up of about 45 $GWd{\cdot}t^{-1}$ with an initial enrichment of 3.7% in U235. The current MOX fuel with an initial Pu content of 7%, is unloaded after 3 reactor cycles and then reaches an average burn-up of 37 $GWd{\cdot}t^{-1}$, thereby causing a lesser performance of the so-called 'hybrid' core in which UOX and MOX assemblies are combined. The goal to be reached by the year 2004 is parity between UOX and MOX in terms of energy generated and costs. This will lead to an average plutonium content of MOX fuel of 8.65%. On the basis of an annual production of 100 t of MOX, this will make it possible to ensure full parity between the separated Pu and recycled Pu flow.

3. Characteristics of spent MOX

The spent MOX fuel differs considerably from that containing uranium, insofar as it contains 4 to 7 times more plutonium (20 to 30 kg per assembly, according to the Burn Up and the initial Pu content) and 5 to 10 times more minor actinides, americium and curium, (about 4 kg per assembly) the effect of that being a markedly higher residual thermal power. This results in a number of constraints, such as the necessity of a longer cooling time before shipment to La Hague (5 years instead of 2), along with the increase in the underwater storage time of several decades.

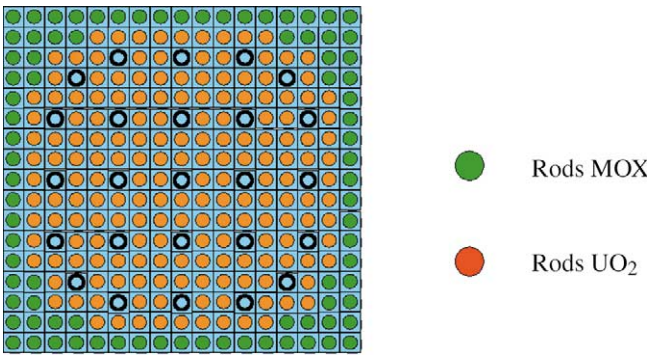

Figure 1. The CORAIL concept.

4. The recycling of plutonium in PWR in the medium term

Considering that French nuclear power plants are comparatively young (the average age being 15 years) and have a useful life expectancy of 40, even 50 or 60 years, and that the next reactor planned, the EPR (European Pressurised Reactor), will also have the same useful life expectation, pressurised water reactors will still exist by the year 2050. During this lifetime of NPPs, it might either be decided to maintain the nuclear option in the long term, so that the Pu would be a necessary resource to be preserved, or to abandon nuclear power generation through a gradual shutdown and dismantling programme, and hence the Pu would become the principal waste one should strive to eliminate.

Plutonium, a high energetic radioelement, is also the main contributor to the so called long-term radiotoxicity of spent fuel, and thus has to be managed in an optimum manner, so as to:

– avoid too great an accumulation of it;
– ensure at the same time that it is available in sufficient quantity and quality to permit the start-up of fast breeders, when the time comes.

With the Pu mono-recycling as practised in the current MOX management system, 20 reactors loaded at 30% with MOX assemblies, the total plutonium inventory in the unloaded and stored MOX assemblies increases annually by about 10 tons.

This major R&D effort is being conducted with EDF-CEA and FRAMATOME-ANP to achieve the goal of optimising the Pu management.

In fact, the first recycling of Pu causes a notable deterioration in the isotopic quality of the residual Pu, which results in a substantial decrease in its energy value with a view to another irradiation in a PWR. Therefore, the multirecycling of this Pu in a standard MOX assembly can hardly be envisaged beyond 2 or 3 recyclings, as the Pu content required to ensure reactivity would be too high with respect to the neutron safety coefficients.

To be able to multirecycle the Pu in PWRs, one has to dissociate the function ‘plutonium mass’ to be fissioned from the function ‘required reactivity’, so as to make it possible to reach the desired burn-up fraction. To this end, one blends at the assembly stage uranium-containing fuel that is enriched with U235, to ensure the reactivity required, with fuel containing plutonium, to provide for the incineration capability required.

On the basis of this process, the French CEA has proposed different assembly designs [2], that is to say:

1. The MIX design, the assembly of which consists exclusively of ‘Pu on enriched U support’, instead of the depleted U of the standard MOX design.

   This assembly design does not pose any particular problem, apart from the extra costs to be provided for, given that its manufacture in large quantities requires plants properly equipped for the handling of Pu.

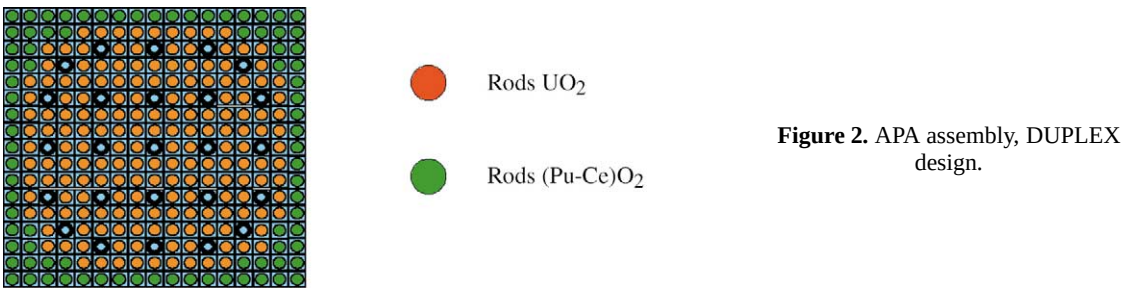

Figure 2. APA assembly, DUPLEX design.

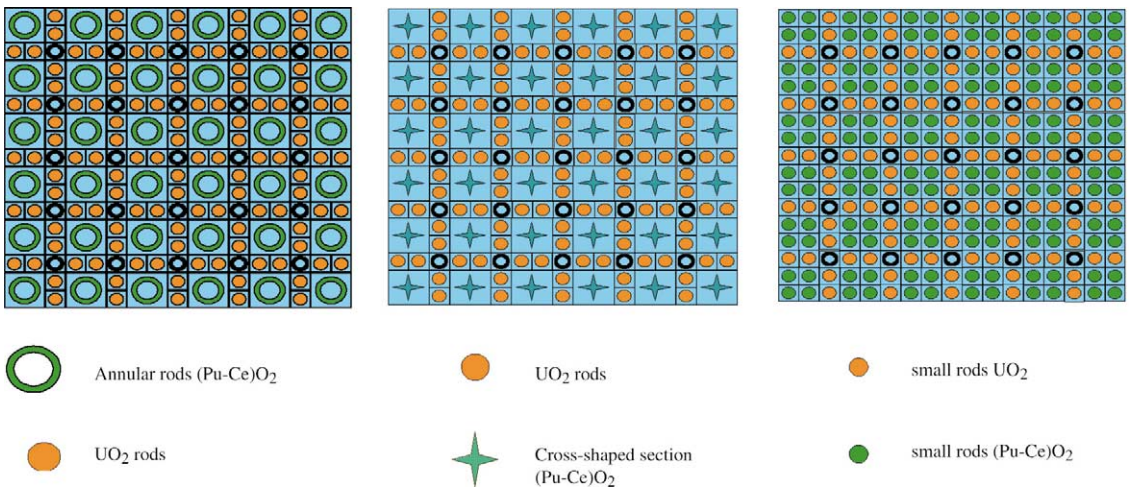

Figure 3. APA assembly of different designs.

2. The 'CORAIL' assembly (Fig. 1) made up of UOX and standard MOX fuel rods, i.e., a bundle of 160 UOX rods surrounded by 84 MOX rods to constitute a shaped conventional assembly and a homogeneous core.
   The U235 content of the UOX rods is adjusted in such a way that the deterioration of the isotopic quality of the Pu is compensated for, to facilitate multirecycling. For example, for a Burn Up of 45 $GWd{\cdot}t^{-1}$ at the 7th recycling, the U235 enrichment must reach 4.80% with a limitation of the Pu content at 8%.
3. The APA (for Advanced Plutonium Assembly) designs, made up of standard UOX rods and of rods with Pu on an inert support without uranium [$(Pu–Ce)O_2$ for instance] within the same assembly; the inert support makes it possible to increase the net consumption of Pu during each irradiation (the inert support does not generate new Pu as depleted uranium support does).

These very innovative designs are organised in two large families (Figs. 2 and 3):

(a) APA made up of 'Pu rods' with standard external geometry, which preserves the moderation ratio, called DUPLEX;

(b) APA with a moderation ratio that is locally increased around Pu rods with optimised geometry: large tubular rods, 'small' rods, large and small cross-shaped sections, the other UOX rods being of the standard type; this design favours a more efficient incineration of the Pu while improving the control of the reactivity of the reactor.

The implementation of these innovative fuels with plutonium, jointly studied by CEA-EDF-COGEMA and FRAMATOME-ANP can be implemented theoretically in the reactors of current NPPs but, this situation is not envisaged. It may be optimised in the European Pressurised Reactors (EPR) that will equip the

future NPPs. In fact, as by its very design, the core of the EPR will feature enhanced flexibility that allows one to envisage, on the one hand, increased moderation effects through design adaptations, and on the other hand, greater adaptability to the future fuel designs than is the case for present PWRs, thanks to the more substantial margins allowed for in the design phase of this new reactor and to the lower fuel power density. With these advanced fuels containing Pu, the stabilisation of the Pu inventory of NPPs producing 400 TWhe·year^{-1} could be achieved within forty years at a level of about 400 t, assuming the use of CORAIL type assemblies, or even of 200 t with APA fuels, but this would produce a Pu considerably more deteriorated for its future use and the price to be paid would be a considerable increase in the quantities of americium and curium produced and sent to waste.

5. Control of the plutonium and minor actinides inventory [3]

To avoid the increasing accumulation of MA (Np, Am, Cm) during the Pu multirecycling, which significantly burdens the benefit expected from Pu incineration in term of radiotoxicity, it is necessary to study the means for their incineration, in order to reduce and then stabilise their inventory in the cycle. The R & D carried out with this respect fall under the scope of the French legislation of 30 December 1991 on long-lived radioactive waste. The work under way, the results of which are expected by 2006, are specifically oriented towards partitioning, transmutation and specific conditioning of the separated radionuclides.

The studies performed in this field are based on the assessment of scenarios that implement three reactor families of the current technology totalising an output power of 60 GWe and generating 400 TWh·year^{-1}:

1. Nuclear generating facilities exclusively made up of PWRs.
2. Nuclear generating facilities exclusively made up of FNRs.

Each of these nuclear generating facilities recycles the Pu and MA in a homogenous manner.

3. A generation mix made up of PWRs loaded with UOX and FNRs loaded with assemblies that recycle Pu and Np as homogenous elements and Am + Cm in dedicated transmutation targets.

Each scenario consists of 3 phases:

- A transitional balancing phase, the starting point of which will be the extrapolated state of current reactors by the year 2010. During this phase, which may last a number of decades, the proportion of reactors incinerating Pu and MA increases as the recycled inventory increases too, until the moment at which the production-to-consumption ratio of the transuranium elements will be balanced within the NPPs.
- A phase of equilibrium that may last as long as the fissile resource is available and that is characterised by the inventory of the transuranics elements in reactor and the reprocessing losses (typically 0.1% for U and Pu, 1% for MA).
- This inventory can be gradually eliminated in a third phase that starts with the shutdown of the producer reactors, is then followed by the reduction of the number of incinerator reactors, as the inventory is incinerated and decreases.

The results of the first detailed studies are illustrated by Figs. 4 and 5 that show, for the scenarios considered, the evolution of:

- the Pu inventory;
- the Am + Cm inventory;

in the various facilities of the whole fuel cycle (reactor, storage facilities, reprocessing plant, waste package…).

These inventories will stabilise after a few decades and result in the equilibrated balances (in metric tons) shown in Table 1.

In these scenarios – by way of comparison with a scenario without reprocessing, in which the entire spent fuel will be disposed of, the reduction of the quantity of transuranium elements contained in the final waste intended for disposal in deep underground repositories is, for 1 TWhe produced at equilibrium, about 130

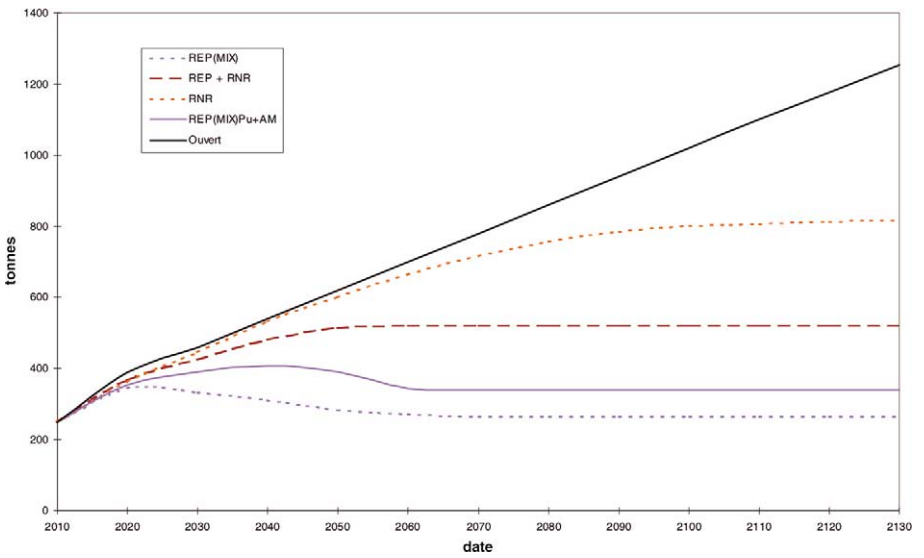

Figure 4. Evolution of the Pu inventory in NPPs (reactors, storage, facilities, waste packages). Ouvert: direct disposal of entire spent fuel; REP (MIX): scenario 1; RNR: scenario 2; REP+RNR: scenario 3.

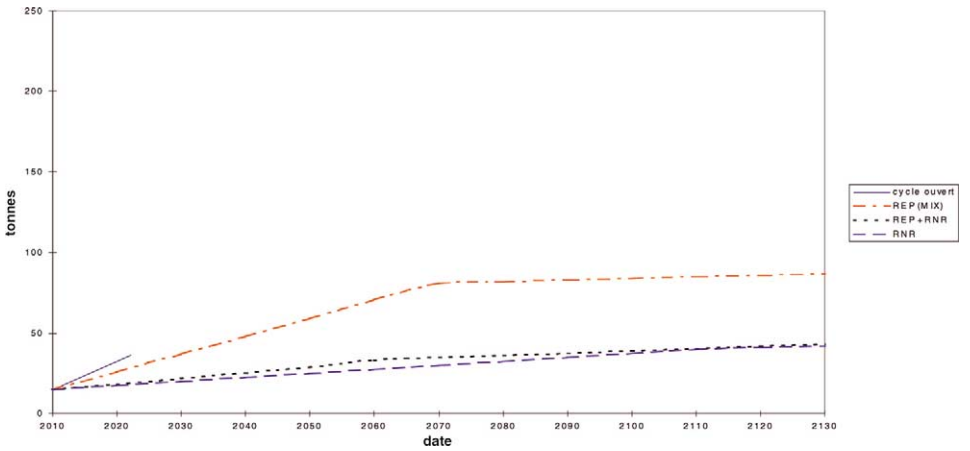

Figure 5. Evolution of the Am + Cm inventory in NPPs (reactors, storage, facilities, waste packages). Ouvert: direct disposal of entire spent fuel; REP (MIX): scenario 1; RNR: scenario 2; REP+RNR: scenario 3.

Table 1. The Pu and MA inventories for the 3 scenarios

	Scenario 1	Scenario 2	Scenario 3
NPPs	100% PWRs (MIX)	100% FNRs	PWRs + FNRs (Am + Cm)
Pu	315	753	471
Am	35.3	31.7	20.8
Cm	48.9	8.1	3.6

for 100% PWRs and 100% FNRs, and 30 to 50 for the generation mix made up of 46% PWRs and 54% FNRs with one recycling of Am + Cm in dedicated transmutation targets.

Though this reduction is lower than for the two first scenarios, the last scenario has the advantage of concentrating the issues related to the handling of the Am and Cm in a minimum of material flows – the targets – and not in all of the fuel of the NPPs.

Lastly, these studies are a theoretical exercise only and do not claim to predict any underlying industrial strategy whatsoever. They are solely intended to provide the information indispensable for the assessment of the pertinence of any of the scenarios above with respect to the industrial realities and the economic background that may prevail at the time considered.

6. Control of the plutonium and minor actinides in innovative systems: example of the molten salt reactor AMSTER [4]

The sustainable development of nuclear power will sooner or later require new reactor types to overcome the limitations that burden the existing systems, no matter whether one examines the issue from the standpoint of the use of natural resources as fuel, of the reduction of long-lived radioactive waste or of economic competitiveness. Among the technological concepts with potentialities, molten salt reactors (MSR) are particularly attractive, thanks to two fundamental features:

- A liquid fuel making it possible to do away with all the constraints related to solid fuel and to achieve considerable simplification of the fuel cycle which, moreover, is fully site-integrated, including, in particular, the possibility of implementing an on-line reprocessing unit through a pyrochemical process, the purpose of which is the extraction of the fission products that poison the core and the feeding with fresh thorium and/or transuranium (TRU) elements to be transmuted.
- The breeding possibility with a thorium cycle and a thermal spectrum. The MSBR (Molten Salt Breeding Reactor) concept proposed by ORNL in the 1970s would thus permit to reach a breeding coefficient of 1.06, with a doubling time of about 25 years, through U233 production.

These two characteristics are particularly interesting with respect to the problem of controlling the Pu and MA inventory: indeed, the TRU elements, which are the main source of long-term radiotoxicity, can be indefinitely recycled in the core until their complete elimination, except for the losses. The latter should be very low thanks to the expected pyrochemical reprocessing performed directly on the molten salt fuel extracted from the core. Furthermore, the thorium support makes it possible to considerably reduce the production of TRU elements, and hence the core inventory is limited by the choice of a thermal spectrum, which, in this case, is another factor in favour of the reduction of reprocessing losses.

The AMSTER (Actinides Molten Salt TransmutER) concept proposed by EDF (Fig. 6) takes maximum advantage of these features to reach the objectives of the main line one of the 1991 legislation by striving to reduce the quantity of salt to be reprocessed per energy unit generated. The aim is to limit as far as possible the size and complexity of the reprocessing unit coupled to the reactor, along with the quantity of TRU elements sent to waste, since the latter is directly proportional to the quantity of salt reprocessed for the extraction of fission products.

This approach led to the abandonment of the breeding objective of the MSBR project, as this objective required reprocessing of the entire core within about ten days, in favour of simple auto-generation of fissile fuel as U233. This has resulted in the definition of a particularly interesting configuration, the core of which is reprocessed in 300 days (i.e., some hundred kg of salt to be processed for a 1 GWe unit). This AMSTER reactor–incinerator of TRU elements could consume (when in balance) 76 kg of thorium and 22 kg of TRU elements coming for instance from spent PWR fuel assemblies, for about 270 g of transuranium elements to be sent to waste per TWhe. The fuel cycle of this reactor is shown in the diagram (Fig. 7) which mentions the different kinds of separation of fission products (FP) (gaseous, metallic, . . .) and the loss ratios at each step of the on-line reprocessing process.

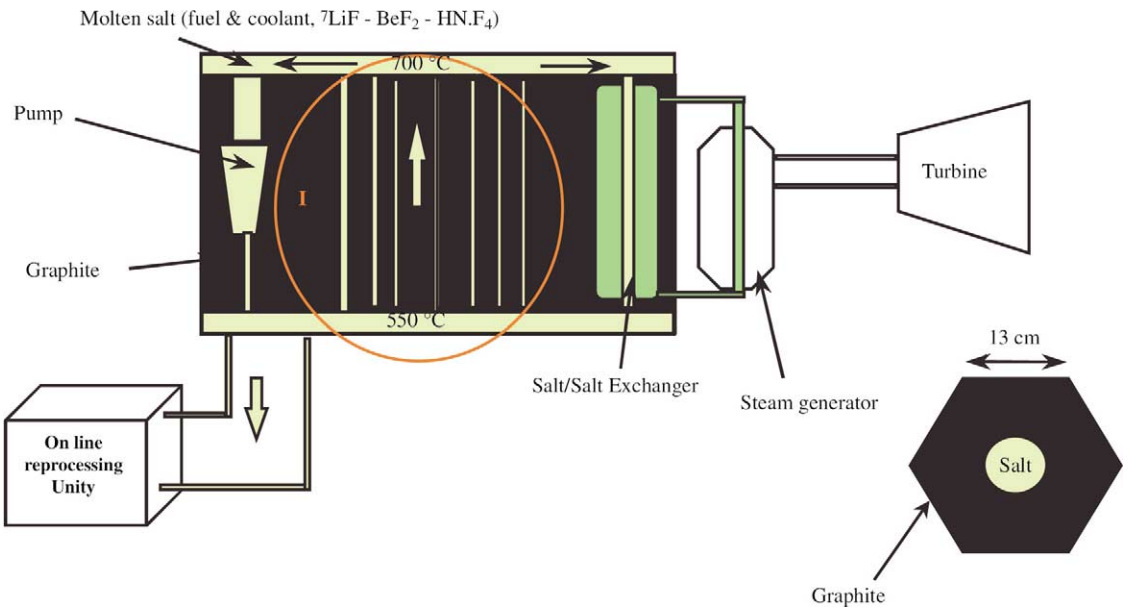

Figure 6. AMSTER: a thermalised molten salt reactor concept.

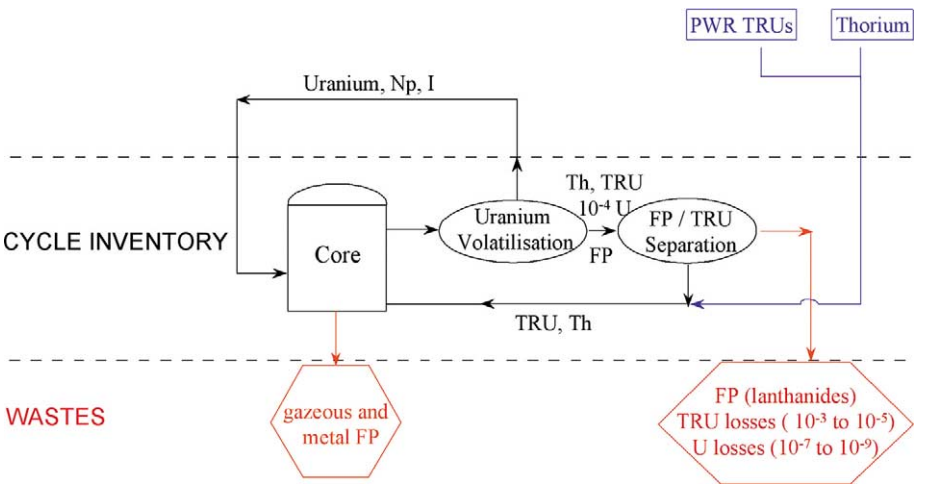

Figure 7. TRU burner on thorium support, fuel cycle.

In addition, this incinerating configuration could serve to produce the U233 necessary for the switch-over to the autogenerating configuration, in which the AMSTER reactor would be loaded with thorium only and function as a U233 autogenerator, so that it would consume 100 kg of thorium for about 20 g of transuranium elements to be sent to waste per TWhe generated.

7. Conclusions

The ambivalent character of plutonium, both a valuable fissile resource in terms of availability of energy resources within the framework of sustainable development and a major contributor to the radiotoxicity

hazard, requires balanced management to develop an autogenerating or breeding system in the medium term while avoiding accumulating too much of it.

The industrial strategy of EDF finds expression in a gradual and sufficiently flexible approach, given the constraining inertia in terms of the time issue of plutonium management, and, more globally, of the back-end of the fuel cycle. The reprocessing of the major part of the spent UOX fuel allows initial recycling of the recovered Pu through the MOX fuels loaded in the 20 licensed reactors while striving towards parity of Pu production and utilisation flows. The storage of spent MOX fuel, containing concentrated Pu makes it possible, eventually, to keep all options open as regards its reprocessing and recovery.

Its possible future multirecycling in new type of reactor such as EPR with respect to the stabilisation of its inventory will then raise the problem of the accumulation of the minor actinides, americium and curium, which will be produced in larger quantities than with conventional UOX assemblies and, as a result, will also confront us with the issue of their long-term management. We then will have to answer the question: "Which plutonium inventory is to be kept for which energy strategy, in particular with respect to nuclear energy long-term strategy?"

The legislation of 30 December 1991 has opened the way for the study of scenarios that might be put into practice for the long-term fuel cycle management, including different possible means for the incineration-transmutation of minor actinides and the optimum use of the fissile resource.

The stabilisation scenarios for the transuranium elements inventories throughout the cycle and the minimisation of the quantity of waste disposed of in deep underground repositories may be supported by:

– either the reactors based on proven technologies, i.e., PWRs, FNRs and technologies derived from these;
– or innovative systems such as hybrid systems, advanced gas coolant reactors or molten salt reactors.

Theoretically, all these technologies with different and unequal levels of maturity should make it possible to reach the set goal. The final choice of a strategy will depend on a number of further criteria, in particular based on the impact on the environment and on health, resistance to proliferation, social aspects, without omitting the overall economy of the systems.

References

[1] International Conference on Back-End of the Fuel Cycle: From Research to Solutions, Proceedings of GLOBAL 2001, Paris, 10–13 September 2001.
[2] J.P. Grouiller, J.Y. Doriath, A. Vasile, J.L. Guillet, D. Greneche, Different possible scenarios for plutonium recycling in PWR's (paper 124).
[3] J.P. Grouiller, G. Vambenepe, B. Carlier, D. Greneche, J. Pavageau, S. Pillon, D. Plancq, C. de Saint-Jean, J. Tommassi, H. Boussier, B. Dinh, J.L. Girotto, A. Lioure, E. Tronche, Waste transmutation scenarios with accessible technologies (paper 122).
[4] J. Vergnes, C. Garzenne, D. Lecarpentier, H. Mouney, M. Delpech, The AMSTER concept: a configuration generating its own uranium (paper 197).

Discussion

Question de J.-M. Loiseaux

Dans l'optique d'un développement du nucléaire au niveau mondial, ce qui correspondrait à un développement de réacteurs à neutrons rapides, un problème apparaît, c'est celui de disposer de suffisamment de Plutonium pour démarrer rapidement une production d'énergie à partir de RNR (U–Pu ou Th–Pu) ; Pouvez-vous commenter sur cette remarque ?

Réponse de H. Mouney

Il est vrai qu'au niveau français, la maîtrise de l'inventaire Pu pourrait se réaliser de manière raisonnée de façon à disposer de Pu en quantité et surtout qualité suffisante (environ 10 t/GWe) pour démarrer une filière rapide surgénératrice ou tout au moins auto régénératrice. Au niveau mondial, le problème sera en fait celui de la ressource fissile disponible au moment où on pourrait décider d'un développement massif du nucléaire en RNR ou autre filière.

Avec les réserves actuelles connues, à coût « raisonnable » (environ 4 à 5 millions de tonnes) et une consommation annuelle de l'ordre de 60 000 à 70 000 tonnes d'uranium naturel, le problème ne se posera pas vraiment avant une soixantaine d'années au niveau mondial. Mais, il est bien évident qu'en cas de redémarrage rapide, il conviendrait de ne pas « gaspiller » la ressource noyau fissile que constitue le plutonium, voire même d'ici là d'amorcer une filière auto génératrice avec le couple Uranium 233–Thorium, possible dans les réacteurs à sels fondus par exemple.

Question de É. Brézin

N'y a-t-il pas une contradiction entre la puissance résiduelle accrue des Mox et l'objectif de réduire à terme les quantités d'actinides ? quelles ont les perspectives de multi recyclage ?

Réponse de H. Mouney

La contradiction n'est qu'apparente puisque la puissance résiduelle accrue des MOX provient du fait que cet assemblage concentre le Pu provenant de 6 à 7 assemblages UOX standard à l'uranium enrichi.

Cet assemblage MOX incinère bien le Pu, mais produit en contre partie 5 à 7 fois plus d'actinides qu'un assemblage UOX.

Le multirecyclage du Pu des MOX est envisageable puisque ces assemblages sont retraitables, mais il n'est pas programmé à court terme car le Pu résiduel des MOX usé est de moins bonne qualité que celui issu des UOX usés et c'est celui-là qu'on utilise en priorité dans les 20 REP 900 MW autorisés pour charger du MOX.

A moyen terme, ce multirecyclage du Pu est étudié dans les réacteurs EPR comme je vous l'ai décrit, au moyen d'assemblages conçus spécialement dans ce but par le CEA, tel que les CORAIL ou APA.